A Study on the Activity Concentration of Po-210 in the Marine Environment of the Kapar Coastal area

(Kajian mengenai Kandungan Po-210 di dalam Persekitaran Marin di Pesisir Pantai Kapar)

Lubna A., Nik Azlin N. A., Afiza Suriani S. & ^{*}Mohamed, C.A.R

School of Environment and Natural Resource Sciences Faculty of Science and Technology Universiti Kebangsaan Malaysia Bangi, 43600, Selangor

*Marine Ecosystem Research Centre (EKOMAR) Faculty of Science and Technology Universiti Kebangsaan Malaysia Bangi, 43600, Selangor

ABSTRACT

The distribution of a natural radionuclide (Po-210) in seawater, total suspended solid (TSS), sediment and marine organisms was quantified using alpha spectrometry at the coastal area of Kapar, Malaysia. It was observed that the Po-210 activity in TSS was much higher than that of water and sediment samples. The ranges of Po-210 activity in organism samples were 4.12±0.11 Bqkg⁻¹ to 7.49±0.14 Bqkg⁻¹ (Arius maculatus), 8.66±0.28 Bqkg⁻¹ to 44.47±0.83 Bqkg⁻¹ (Penaeus merguiensis), 55.59±1.07 Bqkg⁻¹ to 161.68±4.21 Bqkg⁻¹ (Anadara granosa) and 45.70±1.77 Bqkg⁻¹ to 96.44±2.19 Bqkg⁻¹ (Perna viridis). There are significant correlations between the total weight and Po-210 concentration for all the organisms sampled. The concentration factors of Po-210 for the organisms calculated were 1851.42 (Arius maculatus), 14296.58 (Penaeus merguiensis), 95314.84 (Anadara granosa) and 27128.18 (Perna viridis). Therefore, it can be assumed that mollusks can contribute a higher radiation to seafood consumers in the coastal area of Kapar.

Keywords: Po-210, Marine organisms, Concentration factor.

ABSTRAK

Taburan radionuklid semulajadi iaitu Po-210 dalam air laut, jumlah pepejal terampai (TSS), sedimen dan organism marin yang disampel dari kawasan persisiran pantai Kapar dibilang dengan menggunakan spektrometri alfa. Aktiviti Po-210 dalam TSS didapati lebih tinggi berbanding sampel persekitaran yang lain iaitu air laut dan sedimen. Aktiviti Po-210 dalam organism marin pula berjulat dari 4.12±0.11 Bqkg⁻¹ hingga 7.49±0.14 Bqkg⁻¹ untuk ikan duri (Arius maculatus), dari 8.66±0.28 Bqkg⁻¹ hingga 44.47±0.83 Bqkg⁻¹ untuk udang putih (Penaeus merguiensis). Manakala untuk kerang (Anadara granosa) dan kupang ((Perna viridis) berjulat dari 55.59±1.07 Bqkg⁻¹ hingga 161.68±4.21 Bqkg⁻¹ dan dari 45.70±1.77 Bqkg⁻¹ hingga 96.44±2.19 Bqkg⁻¹ masing-masing. Nilai faktor kepekatan Po-210 dalam ikan duri, udang putih, kerang dan kupang masing – masing adalah 1851.42, 14296.58, 95314.84 dan 27128.18. Oleh sebab itu, kajian ini mendapati bahawa moluska iaitu kerang dan kupang mempunyai kadar radiasi yang lebih tinggi berbanding makanan laut lain yang dikaji iaitu ikan duri dan udang putih di kawasan persisiran pantai Kapar.

Katakunci: Po-210, Organisma Marin, Faktor Kepekatan.

Corresponding email: carmohd@ukm.my

INTRODUCTION

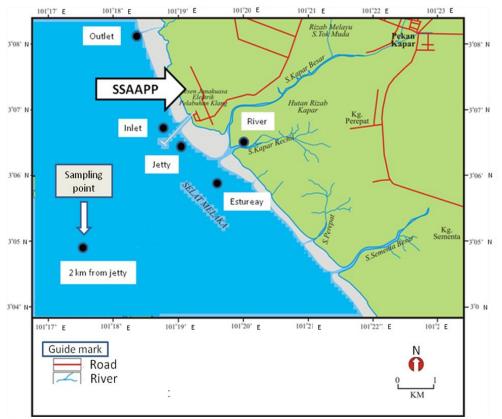
Po-210 ($T_{1/2}$ = 138 d) is a radioactive member of naturally occurring ²³⁸U decay series and the knowledge of the concentration and distribution of this radionuclide is interesting since it provides important information for the monitoring of environmental contamination by natural radioactivity. Po-210 enters the marine environment via the natural radioactive decay of Rn-222 gas, Rn-226 in solution and through wet and dry atmospheric deposition of Bi-210, Pb-210 and Po-210 (Turekian et al., 1977). Po-210 can accumulate in various environmental materials; therefore it can be of great concern from the standpoint of radiation protection because of its radio-toxicity. Po-210 is strongly accumulated by a variety of marine organisms (Cherry and Shannon, 1974) and it is known to be a major contributor to the natural radiation dose received by these organisms (Cherry and Heyraud, 1982). It is also a major contributor to critical group doses from seafood consumption, in particular from the consumption of mollusks (CEC, 1990).

In Malaysia, seafood is considered to be the main source of cheap protein and consumed widely by the population. Therefore, a study regarding Po-210 concentrations in edible seafood is very important for this country in order to assess whether there is an existing health hazard. The main aim of this present study is to monitor Po-210 accumulation patterns in three major groups of seafood; fish, crustacea and mollusks, collected from the Kapar coastal area. The coastal area of Kapar is in the category of a semi-diurnal tidal zone. Sultan Salahuddin Abdul Aziz Power Plant has been operating since 1985 and is adjacent to that coastal area. It is the biggest power plant in Malaysia, contributing 23% of the country's electricity demand, which consumes 2.5 mtpa (million tons per year) of raw charcoal as the main fuel. Therefore, information on Po-210 activities in the marine organisms collected from this area is important for seafood consumers.

MATERIALS AND METHODS

Sampling was carried out on 22th November, 2007, 1st February, 2008, 12th May, 2008 and 27th Aug, 2008, at the Kapar coastal area (03°05′55.4″ N and 101°17′59.7″ E) which is 2 km away from the power plant jetty (Figure 1). Organism samples which were freshly caught from the area around the power plant were also purchased from the local fish market. Approximately 25 L of water samples were collected using polypropylene bottles. Sediment samples were also collected using a Petite Ponar surface grab.

Organism samples were dissected to obtain the edible part (muscle) and oven dried at a temperature of 60°C for 24 hours. Water samples were filtered to separate the Total Suspended Solid (TSS) through pre-weighted Whatman[®] cellulose filter paper (pore size 0.45 μ m). The sediment and TSS samples were also oven dried at 60°C. In all these solid samples, Po-209 tracer was added as a yield tracer and digested with nitric acid and perchloric acid. The solution was filtered and gently evaporated to dryness. Then Po-210 was spontaneously deposited on 2 cm diameter silver disc in 0.5M of hydrochloric acid at 70-90 °C for three hours. The activity of Po-210 was determined by Alpha Spectrometry (Canberra Alpha Analyst, model S570). The filtered water was acidified with concentrated nitric acid (HNO₃) to maintain the pH ≤2. Then about 0.1ml of 25mgl⁻¹ Fe³⁺ as carrier and 0.05 ml of 26.704 dpm ml⁻¹Po-209 as yield tracer were added into the water samples. After that, Na₂CO₃ was added and precipitated with nitric acid and perchloric acid (HClO₄). After heating the solution for 15 minutes, NH₄OH was added to maintain the pH 8 and centrifuged to separate the solution and colloidal in order to obtain solid Fe(OH)₃ precipitate. The precipitated residual was dissolved by HClO₄ and dried at 70 °C temperature. Afterthis, it was dissolved in 80 ml 0.5M HCl and



plated and counted with an alpha spectrometer (Canberra Alpha Analyst, model S570). One way ANOVA and correlation analysis were performed using the Microsoft Excel program for Win XP 2007.

Figure 1: Map Showing the Sampling Point at the Coastal Area of Kapar

RESULTS AND DISCUSSION

Environmental Samples

Po-210 concentrations measured in seawater, TSS and sediment samples are presented in Table 1. Po-210 concentration in water samples ranged from 0.15 ± 0.01 mBql⁻¹ to 0.69 ± 0.02 mBql⁻¹ in February 2008 and May 2008 respectively. In the case of TSS, activity varied from 50.09 ± 1.79 (on February 2008) to 210.37 ±9.39 Bqkg⁻¹ (on May 2008). The concentration of Po-210 in water samples during all the sampling periods maintained a trend similar to that of TSS. Overall, the activities of Po-210 in water are much lower than the TSS. It has already been observed that Po-210 has a strong affinity to suspended particles (Carvalho, 1995). The results of this experiment also agree with that of Connan et al. (2007). In the case of sediment samples, higher activity was found in February 2008 with a value of 21.11 ± 0.94 Bqkg⁻¹ and the lowest activity was observed in May 2008 when the activity was 13.74 ± 0.61 Bqkg⁻¹. This result indicates an opposite trend of Po-210 with the TSS and water samples.

Fish

The results of the study to determine the Po-210 content in fish (*Arius maculatus*) are listed in Table 1. Activities vary from 4.12±0.11 Bqkg⁻¹ (in May, 2008) to 7.49±0.14 Bqkg⁻¹ (in August, 2008). The

highest value was observed in *A. maculatus* with a body weight of 140 g. Apparently these measured values are of a slightly low ranged range when compared to that of coastal fishes from the Kuala Selangor River, which ranges from 0.47 ± 0.23 to 68.10 ± 3.22 Bqkg⁻¹ (Mohamed et al., 2006). This variation may be because of the different habitat, location and feeding behavior of the species. It is has already been proven that the ingestion of food plays a major role in the accumulation of Po-210 (Carvalho and Fowler, 1993). The relationship between Po-210 concentration and total weight of fish was also examined. A good linear correlation (r=0.9166) was observed (Figure. 2a) and there was a significant difference (p<0.05) between total weight and Po-210 concentration. In this study it is clearly demonstrated that the heavier fish accumulates more Po-210 in their muscles. Besides that, Po-210 in this species varies according to the sampling date. Therefore, it can be assumed that Po-210 accumulation is also influenced by environmental factors.

Crustaceans

Po-210 activities measured in *Penaeus merguiensis* ranged from 8.66±0.28 Bqkg⁻¹ in February, 2008 to 44.47 ± 0.83 Bqkg⁻¹ in August, 2008 (Table 1). There is a significant (p<0.05) correlation (r=0.8127) between total weight and Po-210 activities in this species (Figure 2b). Po-210 concentration in shrimps may vary significantly according to the sampling site (Young et al., 2002). Cherry and Heyraud (1981) noted that there appeared to be a steady increase in activity levels when moving from estuarine to coastal and pelagic to deep sea species. In this study, shrimp samples were collected from the local fish market, thus the location is not defined. As a result the variation in Po-210 content may be because of the sampling location and the surrounding environmental conditions. It has been reported that there appears to be a clear relationship between diet and Po-210 concentrations in penaeid and carid shrimps from the north-eastern Atlantic (Heyraud et al., 1988). Shrimps are opportunistic feeders and indiscriminately take food from the benthic zone (Warner, 1977). Some studies have also shown that for the penaeid shrimp, the most important food items are crustaceans, mollusks, polychaetes and other benthic invertebrates (Chong and Sasekumar, 198; Wassenberg and Hill, 1987; Nelson and Capone, 1990; Schmidt, 1993), which are also rich in Po-210. It has already been established that Po-210 fixes easily in organisms such as mollusks and crustaceans because of bonds with sulphur-rich proteins, metallothioneins or ferritin (Durand et al., 1999). Additionally, shrimp can potentially accumulate Po-210 by absorption through the gills or by consumption of contaminated sediment. Therefore, in this study, Po-210 concentration in shrimps is higher than that of fish.

Mollusks

In this study *Anadara granosa* and *Perna viridis* represent the group mollusk. The results of the study to determine the Po-210 content in mollusks are listed in Table 1. The distribution of Po-210 in *A. granosa* varies greatly with the highest value (161.68±4.21 Bqkg⁻¹) recorded in August, 2008 and the lowest value (55.59±1.07 Bqkg⁻¹) observed in November, 2007.In case of *P. viridis* the Po-210 concentration varied from 45.7±1.17 Bqkg⁻¹ to 96.44±2.19 Bqkg⁻¹ (Table 1). This present study recorded a higher level of Po-210 in *A. granosa*. This species has contact with sea sediments and this habit may contribute to the higher levels of Po-210. On the other hand, *P. viridis* lies attached to wharf pilings, sea walls and rocks (Barnes, 1974) and hence does not have direct physical contact with sediments, accounting for a relatively lower level of Po-210 accumulation. It is unlikely that the differences between these two bivalves, in term of Po-210 concentration, arise from the feeding process, as they are filter feeders and feed by straining suspended matter and food particles from water. The differences in accumulation ability might be affected by their ecological niche and environmental factors in the sampling area. Among all the organisms analyzed in this study, mollusks showed greater accumulative capability than shrimp and fish. This result also agrees with that of Shaheed, et al. (1997) who studied the distribution of Po-210 in the abiotic and biotic components of

the Kaveri river ecosystem. It has been found that Po-210 activities and total weight are significantly (p<0.05) correlated for both species. (Figure 2 c,d,).

Connan et al. (2007) proved that mussels that are heavier have a higher Po-210 activity. The same pattern of Po-210 accumulation was also found in this study.

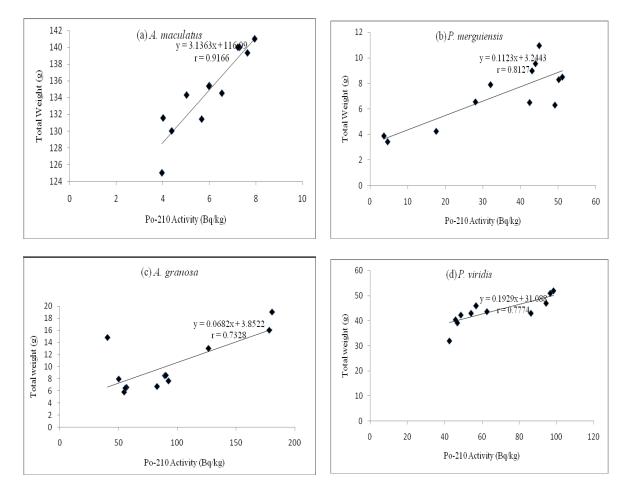


Figure 2: Correlations Between Po-210 and Total Weight of Organisms.

Concentration factors

Concentration factors are used as transfer parameters in assessments of the public dose of radioactivity in the marine environment (Tateda and Koyanagi, 1996). It is defined as the ratio of the activity in species in Bqkg⁻¹ (w.w) to the activity in seawater (filtered at 0.45 μ m) in Bql⁻¹ (Connan et al., 2007). In this study the concentration factors (CFs) were calculated on the basis of the values for Po-210 activities measured in organism and water samples. In this case the following equation was used,

CF = Activity in wet weight of specified tissue sample (Bq/kg)/Activity in filtered water (Bq/l) (Tee, 2004)

Sample	22 th November, 2007		1 st February, 2008		12 th May, 2008		27 th Aug, 2008	
	Total	Po-210 activities	Total	Po-210	Total	Po-210	Total	Po-210
	weight (g)	(Bq/kg)	weight (g)	activities	weight (g)	activities	weight (g)	activities
				(Bq/kg)		(Bq/kg)		(Bq/kg)
Arius maculatus	135.42	6±2.52	139.32	7.64±0.43	131.55	4.01±0.18	140	7.3±0.21
	135.35	5.98±2.51	134.51	6.54±0.5	125	3.96±0.18	140	7.23±0.32
	134.29	5.02±2.52	131.41	5.67±0.6	130	4.38±0.2	141	7.95±0.15
Mean	135.02	5.67±1.45	135.08	6.62±0.3	128.85	4.12±0.11	140.33	7.49±0.14
Penaeus merguiensis	8.99	43.09±0.97	3.42	4.68±0.21	6.55	28.04±1.25	7.9	32.05±1.15
	10.97	45±0.5	3.88	3.7±0.16	6.3	49.17±2.19	8.5	51.15±1.95
	9.55	44±0.9	4.24	17.6±0.79	6.5	42.44±1.89	8.3	50.2±1.06
Mean	9.84	44.03±0.47	3.85	8.66±0.28	6.45	39.88±1.05	8.23	44.47±0.83
Anadara granosa	6.57	56.59±1.67	8.5	89.2±5.99	14.8	40.38±1.8	13	126.37±5.64
	5.8	54.581.8	6.72	82.55±7.65	7.63	92.31±4.12	16	178.14±7.95
	6.45	55.59±2.05	8.55	90.09±6.46	7.95	49.95±2.23	19	180.52±8.05
Mean	6.27	55.59±1.07	7.92	87.28±3.89	10.13	60.88±1.67	16	161.68±4.21
Perna viridis	40.4	45.83±2.64	43	54.16±2.42	43	86.26±3.85	51	96.62±3.36
	42.28	48.69±1.17	46	56.75±2.53	39.14	46.82±2.54	47	94.37±3.36
	31.94	42.55±1.99	46	56.85±2.54	43.66	62.67±2.8	52	98.34±4.02
Mean	38.21	45.69±1.17	45	55.92±1.44	41.93	65.25±1.8	50	96.44±2.19
Sediment		20.33± 0.33		21.11±0.94		13.74±0.61		14.31± 0.64
TSS		149± 2.23		50.09±1.79		210.37±9.39		75.56± 3.37
Water (mBq/l)		0.55± 0.02		0.15±0.01		0.69± 0.02		0.51 ± 0.04

Table1: ²¹⁰Po concentration in organisms, Sediment, TSS and Water samples during different sampling periods.

For this calculation the values of activities in the seawater of each sampling period $(0.55\pm0.02 \text{ mBql}^{-1} \text{ in November, 2007; } 0.15\pm0.01 \text{ mBql}^{-1} \text{ in February, 2008; } 0.69\pm0.02 \text{ mBql}^{-1} \text{ in May, 2008 and } 0.51\pm0.04 \text{ mBql}^{-1} \text{ in August, 2008}) were used. Generally, higher concentrations are seen for mollusks (95314.84 in$ *A. granosa*and 27128.18 in*P. viridis*). The concentration factor values for another two groups of species are 1851.42 and 14296.58 in fish and crustaceans respectively. Therefore, among the three groups of marine organisms, mollusks may be expected to contribute a higher exposure to man via dietary intake. In the case of fish and crustaceans, observed concentration factor values obtained in this study were lower than the values published by the IAEA (1985) which are 2000 for fish, 50,000 for crustaceans. But the CF value of*A. granosa*calculated in this study (95314.84) crossed the IAEA standard (10,000 for mollusks). On the other hand the CF of*P. viridis*(27128.18) is within the range. Mohamed et al. (2006) also found the lower CF values in fish species caught form the coastal waters of Kuala Selangor in Malaysia. Lower concentration factors for fish compared to mollusk tissues were observed from the same study area (Tee, 2004). Therefore, it can be concluded that among the seafood items in Malaysia, mollusks are considered to be the higher contributor of Po-210 exposure to humans as a result of seafood consumption.

CONCLUSION

This study provides a general view of α -emitting radionuclide Po-210 in the coal fired power plant area. Po-210 was non-uniformly distributed with the groups of organisms. The higher values were associated with mollusk and lower values with fish. The middle position was occupied by crustaceans. The different levels of Po-210 in different kinds of seafood were also due to species habitats. This study revealed the correlation between an organism's total weight and Po-210 accumulation ability. Therefore it can be assumed that Po-210 was accumulated through the food chain. Calculated concentration factor values were compared with the reported values of IAEA and comparatively lower concentration factors were found in fish and crustaceans but higher values are demonstrated in mollusks. Thus, it can be expected that a higher radiation dose can be received through the dietary intake of mollusks at the Kapar coastal area.

ACKNOWLEDGEMENT

The authors would like to thank all the individuals and lab members involved in this research, especially Asnor, for his cordial help during sampling. We would also like to acknowledge the help of Kapar Energy Ventures Sdn. Bhd. This research is supported by the grant of NOD/RED/02/001.

REFERENCES

Barnes, R.D., 1974. Invertebrate zoology. W.B. Saunders Company, Philadelaphia, PA, pp. 807

- Carvalho, F.P., 1995. ²¹⁰Pb and ²¹⁰Po in sediments and suspended matter in the Tagus estuary, Portugal. Local enhancement of natural levels by wastes from phosphate ore processing industry. *Sci. Total Environ.* **159**, 201–214.
- Carvalho, F.P., Fowler, S.W. 1993. An experimental study on the bioaccumulation and turnover of polonium-210 and lead-210 in marine shrimp. *Mar. Eco. Prog. Ser.* **102**, 125-133.

- CEC., 1990. The radiological exposure of the population of the European Community from radioactivity in North European marine wasters- Project 'Marina'. Commission of the European Community. Report No. EUR 12483, Luxembourg.
- Cherry, R.D., Heyraud, M., 1981. Polnium-210 content of marine shrimp: Variation with biological and environmental factors. *Mar. Biol.* **65**, 165-175.
- Cherry, R.D., Heyraud, M., 1982. Evidence for high natural radiation doses in certain mid-water oceanic organisms. *Science*. **218**, 54–56.
- Cherry, R.D., Shannon, L.V., 1974. The Alpha Rradioactivity of Marine Organisms, *Atom. Energy Rev.* **12**, 3–45.
- Chong, V.C., Sasekumar, A., 1981. Food and feeding habits of the white prawn *Penaeus merguinsis*. *Mar. Ecol. Prog. Ser.* **5**, 185-191.
- Connan, O., Germain, P., Solier, L., Gouret G. 2007. Variations of ²¹⁰Po and ²¹⁰Pb in various marine organisms from Western English Channel: contribution of ²¹⁰Po to the radiation dose. *J. Environ. Radioac.* **97(2-3)**, 168-88.
- Durand, J. P, Carvalho, F.P, Goudard, F., Pieri, J., Fowler, S.W., Cotret, O. 1999. ²¹⁰Po binding to metallothioneins and ferritin in the liver of teleost marine fish. *Mar. Ecol. Prog. Ser.* **177**, 189–196.
- Heyraud, M., Domanski, P., Cherry, R.D., Fasham, M.J.R. 1988. Natural tracers in dietary studies: data for ²¹⁰Po and ²¹⁰Pb in decapods shrimp and other pelagic organisms in the Northeast Atlantic Ocean. *Mar. Biol.* **97**, 507-519.
- IAEA, 1985. Sediment K_ds and concentrations factors for radionuclides in the marine environment. Tech. Rep. Series No. 247, Vienna, Austria.
- Mohamed, C.A.R., Tee, L.T., Zal, U. M., Zaharuddin, A., Masni, M. A. 2006. Activity concentrations of ²¹⁰Po and ²¹⁰Pb in edible tissue of fish caught at Kuala Selangor, Malaysia. *Malays. Appl. Biol.* **35(2)**, 67-73.
- Nelson, W.G., Capone, M. A., 1990. Experimental studies of predation on polychaetes associated with seagrass beds. *Estuaries.* **13**, 51-58.
- Schmidt,T.W., 1993. Community characteristics of dominant forage fishes and decapods in the whitewater Bay-Shark River Estuary, Everglades. Everglades National Park Technical Report 93/12, US National Park Service, Atlanta, Geoggia.
- Shaheed, K., Somasundaram, S. S. N., Hameed, P. S, Iyengar, M. A. R. 1997. A study of Polonium-210 distribution aspects in the riverine ecosystem of Kaveri, Tiruchirappalli, *India. Envron. Pollution.* **95(3)**, 371-377.
- Tateda, Y., Koyanagi, T., 1996. Concentration factors for ¹³⁷Cs in Japanese Coastal fish (1984-1990). *J.Radiat. Res.* **37**,71-79.
- Tee, L. T., 2004. Activities of Polonium-210 and Lead-210 in marine environments of Kuala Selangor, Pulau Besar and Pulau Redang. MSc Thesis, Universiti Kebangsaan Malaysia.
- Turekian, K. K., Nozaki, Y., Benninger, L. K., 1977. Geochemistry of atmospheric radon and radon products. *Ann Rev. Earth Planet Sci.* **5**, 227-255.
- Warner, G. F., 1977. The Biology of crabs, Paul Elek (Scientific Books). London.
- Wassenberg, T. J., Hill, B. J., 1987. Natural diet of the tiger prawns *Penaeus esculentus* and *P. semisulcatus. Aust. J. mar. Freswat. Res.* **38**, 169-182.
- Young, A. K., McCubbin, D., Camplin, W. C., 2002. Natural Radionuclides in Seafood. Environment Report RL17/02.